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Anomalous Elevated Radiocarbon Measurements of $PM_{2.5}$

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Two-component models are often used to determine the contributions made by fossil fuel and natural sources of carbon in airborne particulate matter (PM). The models reduce thousands of actual sources to two end members based on isotopic signature. Combustion of fossil fuels produces PM free of carbon-14 (^{14}C). Wood or charcoal smoke, restaurant fryer emissions, and natural emissions from plants produce PM with the contemporary concentration of ^{14}C approximately 1.2 $x10^{-12}$ $^{14}C/C$. Such data can be used to estimate the relative contributions of fossil fuels and biogenic aerosols to the total aerosol loading and radiocarbon analysis is becoming a popular source apportionment method. Emissions from incinerators combusting medical or biological wastes containing tracer ^{14}C can skew the $^{14}C/C$ ratio of PM, however, so critical analysis of sampling sites for possible sources of elevated PM needs to be completed prior to embarking on sampling campaigns. Results are presented for two ambient monitoring sites in different areas of U.S. where ^{14}C contamination is apparent. Our experience suggests that such contamination is uncommon but is also not rare (~ 10%) for PM sampling sites.

Introduction

Two component end member models are often used for source apportionment studies of particulate matter (PM) to estimate the contributions of major carbon sources [1-11]. Carbon-14 (^{14}C) can be used to distinguish between fossil carbon free of ^{14}C and biogenic carbon possessing a contemporary $^{14}C/C$ ratio. The fossil carbon component is produced primarily from fossil fuel combustion, most commonly assigned to diesel engines and large stationary sources such as coal-fired power plants. The biogenic component is a mixture of natural processes and anthropogenic activities. Biomass burning from wild fires, residential wood burning and campfires, secondary organic aerosols (SOA) from vegetation, and food cooking produce high levels of PM with carbon that generally reside in the atmosphere for 3-7 days. Significant biogenic carbon has been reported in PM retained on a 2.5 μm filter ($PM_{2.5}$) and on smaller particles [1-11]. The high biogenic carbon content of PM in these studies was generally attributed to SOA whether they were collected at remote, rural or metropolitan sites. Increased PM due to seasonal residential wood burning was the focus of two studies [3,8]. Bench and Herckes [1] applied source apportionment to the impact of haze in Yosemite National Park in California, USA. They correlated an increase in haze with an increase in biogenic PM produce by forest fires hundreds of kilometres away from the sampling site [1]. At rural sites studied, the fossil component of the PM did not vary much with the season. The fossil component of the PM was larger in the winter at the urban sites. The fossil component of PM was greater urban sites than rural sites, but the biogenic component was still usually larger [4,7,10,11]. Most of the variation in PM filter loading was due to swings in biogenic carbon contribution.

Occasionally PM filters are collected with $^{14}C/C$ ratios elevated significantly above the contemporary level and in some cases above the peak of the bomb pulse. The elevated $^{14}C/C$ is due to combustion or emission of tracer ^{14}C that is integrated into or deposited onto the PM, producing super modern PM. Our experience suggests that such super modern ^{14}C contamination is uncommon, but is also not rare (~10% of sites have some elevated samples). Other researchers have measured super modern PM samples, but these results are not reported. Sites that produce super modern PM samples are generally abandoned after measurement of the first hot sample. A medical waste incinerator was capable of producing a super modern forest outside Oak Ridge, TN USA [12-14] and releases from nuclear facilities and radioisotope laboratories were implicated in producing locally elevated vegetation [15-17]. All medical waste and low-level radioactive waste incinerators are potential sources of tracer ^{14}C in PM since isotopic signatures are carried from fuel through the combustion process to PM [18]. Additionally, hood exhaust from laboratories using ^{14}C tracer can sorb onto existing particulates to produce elevated PM. PM has very high surface area, providing many sites for adsorption of volatile carbon molecules. This property is employed in fullerene soot aerosol monitors deployed in and around AMS facilities [19,20]. Hence, ambient PM can adsorb elevated $^{14}CO_2$ from incineration of low-level radioactive waste or tracer ^{14}C emissions from fume hoods.

Concentrations of $^{14}C/C$ in $PM_{2.5}$ are presented for two ambient monitoring sites in different areas of U.S. where ^{14}C contamination is apparent. The sites were more than 2000 km apart, rural central Illinois and Los Angeles, California, and samples were processed independently. Different sampling equipment was used at each site.

Potential sources of elevated ^{14}C that led to the super modern PM samples are identified based on potential incineration and ^{14}C tracer sources and wind patterns. We present results intended as a warning regarding uncritical interpretation of ^{14}C results, particularly when $^{14}C/C$ levels are within the expected range and low-level contamination would not be obvious.

Methods

Bondville, Illinois site

The first site with super modern PM samples was the Bondville Environmental and Atmospheric Research Site (BEARS) (elevation 215 m, nominal pressure 741 mm Hg). The BEARS location was a rural site in Champaign County, Illinois USA, located ~ 13 km southwest from Champaign-Urbana, Illinois, home of the University of Illinois with a population of about 210,000 people (Fig. 1). Aside from Champaign-Urbana, Bloomington 80 km northwest, and Decatur 70 km southwest of Bondville, there are no moderately sized cities within 120 km. The terrain is very flat and dominated by corn and soybean fields. Bondville is located 220 km south of Chicago, 200 km west of Indianapolis, and 290 km east of St. Louis. The BEARS site has meteorological instrumentation and has been used for other PM studies [21-23]. The site is a 5.72-hectare rectangular (220 meter by 260 meter) plot in a central part of a 102-hectare farm owned and managed by the University of Illinois Foundation. Sampling was conducted during summer 2004 as part of an IMPROVE (Interagency Monitoring of Protected Visual Environments) program to investigate the carbon-14 content of the collected particles at various IMPROVE sites [4].

Wilmington, California site

Wilmington is a district of the city of Los Angeles adjacent to the Port of Los Angeles and near the Port of Long Beach (Fig. 2). Wilmington is an urban, industrial setting with significant diesel traffic from ships, trains, and trucks. It is also home to multiple oil refineries. A couple of studies of air pollution in Wilmington have been published [24,25] due to the high diesel traffic, but there is not a long standing monitoring station like Bondville. The sampling site was at an elementary school 3 km north of the Port. An earlier Children's Environmental Health Protection study in Wilmington by the California Air Resources Board is available online [26].

Sampling

Sampling at Bondville was conducted during a three-month summer period (June, July, August) in 2004. Each week, sampling began at midnight Tuesday night/Wednesday morning and ended at midnight Monday night/Tuesday morning. Tuesday was a non-sampling day to allow the filters to be changed during the regular IMPROVE sampler visit. Sampling at Wilmington was conducted for the entire year of 2007.

Samples were collected using a Thermo-Anderson High Volume Sampler (Hi-Vol) model GS2313-105BL with a brushless motor and a 2.5 μm cut point impactor (model SA231-F). This impactor plate gives a $PM_{2.5}$ sample on the backup filter at 18.9 volumetric L/s. Flow for this sampler is controlled by a mass flow controller and the sampler was adjusted to operate at a volumetric flow of 18.9 L/s. At the altitude of the BEARS site 18.9 volumetric L/s translates to an 22.6 g/s mass controlled flow rate.

One slotted glass fibre substrate was installed in the TSP Hi-Vol impactor head per sampling period to collect particles greater than 2.5 μm when the sampler operated at 18.9 volumetric L/s. These substrates were only used for sizing and were discarded following sampling. One quartz fibre filter per sampling period was used to collect particles 2.5 μm and below at 18.9 volumetric L/s. Quartz fibre filters 20x25 cm (Gellman QM-A) were pre-fired by baking at 600 °C overnight (about 12 hours) before use. Filters were sealed in clean plastic bags before and after sampling to reduce exchange with the ambient air.

Once a month, on the Tuesday nearest the middle of the month, a quartz filter field blank was exposed. The blank quartz filter was removed from its plastic bag, mounted in the sampler as you would any normal filter, and left in the sampler for ten minutes with the pump off. The quartz filter was removed from the sampler and placed in a zip-locked plastic bag.

Following sampling, the quartz filters were sealed in plastic bags and shipped to LLNL for analysis Upon receipt at LLNL individual filters were logged and stored in a cool dry environment prior to sample preparation. Samples were prepared and analyzed for total aerosol carbon loading and ^{14}C content as previously described [1] with the exception that up to 10x10 cm (100 cm^2) areas of the quartz fibre filters were analyzed. AMS analyses used QA/QC procedures as previously described [1,4].

Results and Discussion

Bondville

The $^{14}C/C$ ratios of the weekly filters are shown in Figure 3. About half of the Bondville filters possessed super modern $^{14}C/C$ ratios indicating tracer contamination. All filters were resampled and the elevated results were repeated. Filters from other IMPROVE sites processed concurrently did not possess excess ^{14}C. Carbon loading on the filters was compared to ^{14}C level (Figure 4). Although the filter with the highest ^{14}C also had the highest carbon loading, there was no correlation between $F^{14}C$ and μg carbon/m^3 of air.

Based on knowledge of the labelled forest produced by the medical waste incinerator, we looked for potential combustion sources of ^{14}C tracer in the vicinity of BEARS. BEARS is in the midst of a cornfield, not close to any combustion source or known ^{14}C tracer facility. Throughout the year two small incinerators at the University of Illinois at Urbana-Champaign Champagne campus 13 km northeast from the Bondville site dispose of experimental plant and animal materials that may contain ^{14}C at tracer levels. Because their total emissions are well below the applicable reporting limits, neither incinerator was included the 1999 National Emissions Inventory listing of point sources. The larger facility is the Veterinary Medicine incinerator that incinerates every Thursday at a minimum. The smaller facility's operation schedule is unknown over the sampling period. There are also numerous laboratories throughout the University campus that use ^{14}C tracer.

We looked through meteorological records at BEARS to determine wind direction during the weeks that had elevated PM samples. The prevailing wind

direction in the summer months is west to southwest with northeast winds relatively uncommon at Bondville in the summer months. A medical waste incinerator in Clinton is generally upwind of BEARS, but 50 km away. The city of Decatur is upwind of BEARS through most of the summer, but also too far away (70 km) to be the likely source of ^{14}C. The University sources (incinerators, tracer labs, coal-fired power plant) are generally downwind of BEARS, but only 13 km away. Figure 1 shows the distribution of likely sources of the elevated ^{14}C at BEARS. In Figure 3 the number of 6 h intervals during which the wind blew from Champaign-Urbana to BEARS is depicted with arrows. Although not a perfect correlation, the elevated filters did experience a prolonged period of suitable wind direction to deposit elevated PM generated at the University at BEARS. Two weeks with favourable northeast winds did not produce super modern PM, but the ^{14}C sources may not have been operating during those wind conditions.

Wilmington

The distribution of $^{14}C/C$ in PM from Wilmington over the year is shown in Figure 5. Through most of the year the $F^{14}C$ is about 0.5. It is dominated by diesel exhaust and on-shore wind flow. During November and December there are several filters with super modern ^{14}C. Unlike Bondville, there was no meteorological station associated with the Wilmington site. Although detailed wind direction data of the sampling site is not available, general wind trajectories for the LA basin are available online [28]. Offshore winds were common during the time of the super modern PM collection. Winds blow across the Los Angeles basin and out to sea, carrying PM and volatile compounds produced or released in Los Angeles past Wilmington. There are

numerous incinerators and commercial users of ^{14}C tracer in the Los Angeles basin. The National Emission Inventory lists 1076 emission sources for $PM_{2.5}$ in Los Angeles county in 2002 [29]. There are also companies that use and produce ^{14}C-lableled molecules for research in the vicinity. PM samples collected at other sites in the Los Angeles basin and measured in our lab also saw sporadic super modern PM samples during the November-December 2007 time frame. The limited detail of wind trajectory information and multitude of potential sources of elevated ^{14}C prevented identification of specific sources for tracer ^{14}C in Wilmington.

Conclusion

Super modern $PM_{2.5}$ samples are uncommon, but not rare. Over the past 12 years we have seen unnaturally elevated ^{14}C levels in PM in at least some samples from about 10% of the sites surveyed. Super modern samples were obtained in both urban and rural settings. Even a remote site such as the San Gorgonio Wilderness 125 km east of Los Angeles on the east side of the San Bernardino Mountains has produced elevated PM samples. A survey of vegetation at San Gorgonio did not detect elevated ^{14}C, so super modern material on the PM occurred too briefly to be recorded by the plant growth or it was not present in CO_2 incorporated into the plants. It is not possible to tell if only some of the samples at a site saw elevated PM. If the increase is subtle, the shift in $F^{14}C$ may keep the PM less than contemporary. These problem sites should be avoided. Generally one or more potential sources of elevated ^{14}C PM can be identified after filters are acquired. Ideally, researchers want to avoid problem sites before starting a sampling campaign.

Avoiding sites near medical waste or low-level radioactive waste incinerators is mandatory. Identification all incinerators in the area, both big and small, should be completed before selecting a site. The National Emission Inventory (NEI) maintained by the U.S. Environmental Protection Agency provides lists and maps of PM sources [30] that can be queried by geography, air toxin, and industry. The NEI list is dated and may not include current information. Typically licenses regulated by state or local agencies are required for operation of incinerators. Information on licensees in an area is available from the appropriate government agency. Conducting web searches for business or facilities engaged in incineration, waste processing, biological waste disposal, radioactive waste disposal, or similar topics can locate potential emission sources.

Since PM from combustion sources tends to have high surface area it can adsorb molecules from aerosols. It is possible that any volatile, labelled compound can contribute to elevated PM samples. The fullerene soot mixed with iron powder that we use to monitor aerosols in AMS prep labs is essentially PM packed into an AMS target [19,20]. The lab aerosol monitors detect ^{14}C releases as subtle as defrosting freezers holding archived tissues from ^{14}C bioAMS studies [19]. We have measured elevated aerosol monitors and connected it to the use of ^{14}C-benzene in a building near our preparation labs [19]. Although PM can clearly adsorb a variety of organic molecules, it is not clear if ^{14}C-labeled molecules expelled by fume hoods can have an impact outside the immediate vicinity of the source. The labelled forest in Oak Ridge, Tennessee serves as a warning that large amounts of $^{14}CO_2$ can be released from incinerators. One often cannot know *a prior* if a small combustion source will

contaminate samples. The techniques described above to find PM emission sources can also be applied to identify users of radiotracers near a proposed sampling site. Keeping PM sampling sites well separated from known users of ^{14}C tracer and incinerators is the best tactic when selecting sampling locations.

Despite researching a site in detail, PM samples can still be contaminated by an unidentified source. To minimize the likelihood of this, it is best to conduct a brief field test prior to embarking on a long sampling campaign. Also, it is better to analyze some PM samples quickly rather than waiting for the completion of a seasonal or annual collection campaign before conducting analyses. If a ^{14}C tracer source is identified after the start of sampling, plume transport modelling might be able to salvage some of the data if detailed meteorological information is available. Using concurrent molecular analyses or compound class analyses or separations in conjunction with radiocarbon might indicate a specific fraction of PM is problematic. Selecting sampling sites free of super modern carbon sources is always the better approach.

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Figure Captions

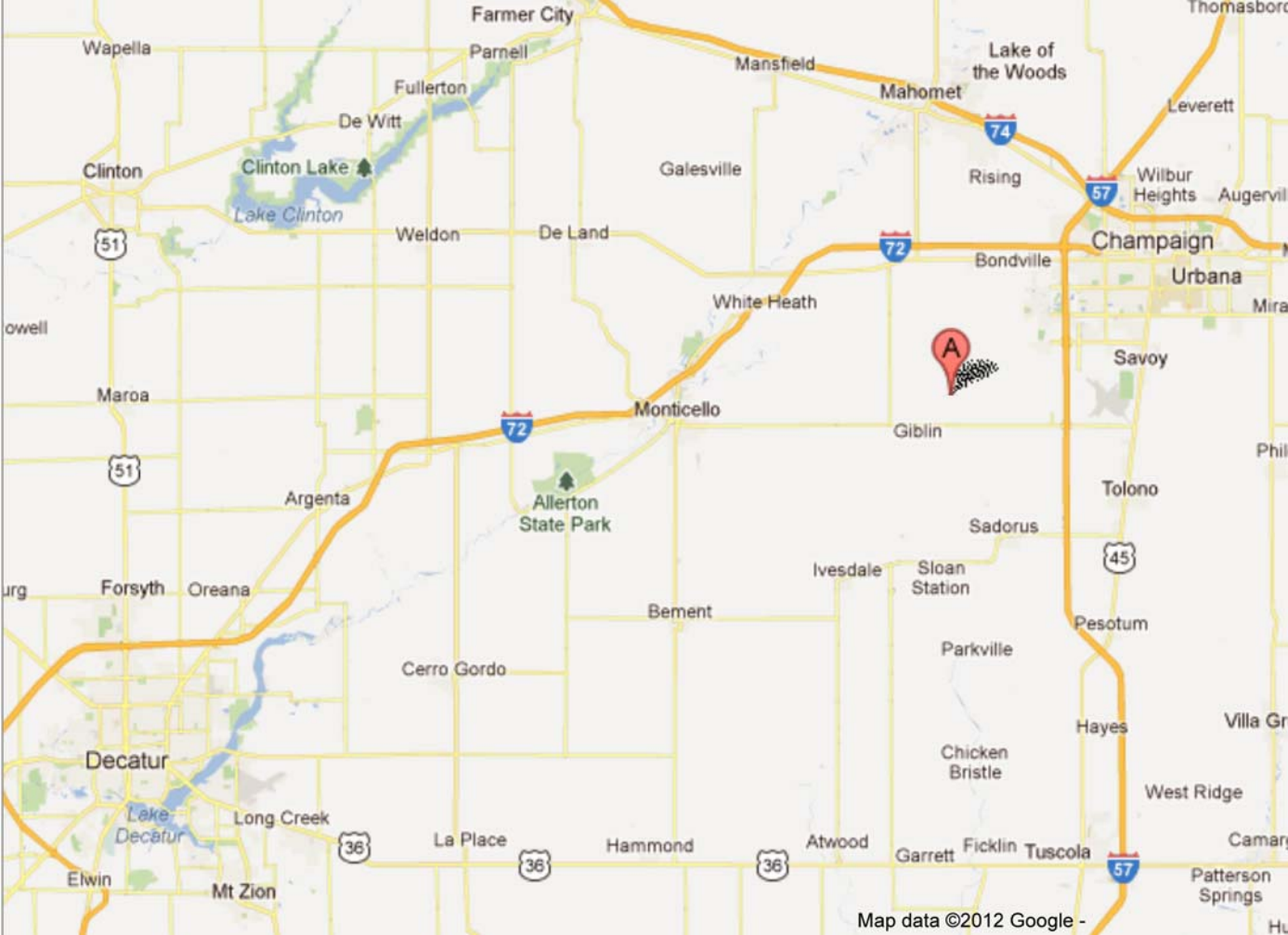
Figure 1. Map of the BEARS site (denoted by A) and likely sources of super modern PM. A medical waste incinerator was located in Clinton 50 km west of BEARS and generally upwind during sampling. Decatur, 70 km southwest of BEARS, is also generally upwind. The University of Illinois at Urbana-Champaign is 13 km northeast of BEARS and generally downwind. The University operated two incinerators that combusted animal and plant research waste, a coal-fired electrical generating plant, and numerous laboratories using ^{14}C tracer. Map is provided courtesy of Google Maps.

Figure 2. Map of the Wilmington District sampling site (denoted by A) in Los Angeles. The Ports of Los Angeles and Long Beach are 3 km south of Wilmington. More than 1000 point sources of $PM_{2.5}$ are listed for Los Angeles County [29]. There are too many potential sources of the elevated ^{14}C from combustion or fume hood emissions to identify likely sources. In addition to general industrial activity and large research universities in Los Angeles, custom radioisotope synthesis companies are found with 40 km north and east of Wilmington.

Figure 3. Carbon-14 concentrations of particulate matter in $F^{14}C$ [27] at Bondville on filters collected weekly. The solid horizontal line denotes the atmospheric level of $^{14}C/C$ at the time of sampling. Each vertical arrow above a weekly histogram signifies 6 continuous hours of wind direction from the University campus to BEARS. Error bars denote 1 standard deviation.

Figure 4. Carbon mass loading on filters compared to $F^{14}C$ level. The solid horizontal line denotes the atmospheric level of $^{14}C/C$ at the time of sampling. Error bars denote 1 standard deviation. Field blanks had $F^{14}C = 0.000 \pm 0.003$ with loading 0.00 ± 0.05 $\mu g/m^3$ [4].

Figure 5. Carbon-14 levels in $F^{14}C$ on the filters collected in Wilmington. The contemporary $F^{14}C$ level of the atmosphere during collection is shown with the nearly horizontal line. Error bars denote 1 standard deviation and are smaller than the symbols.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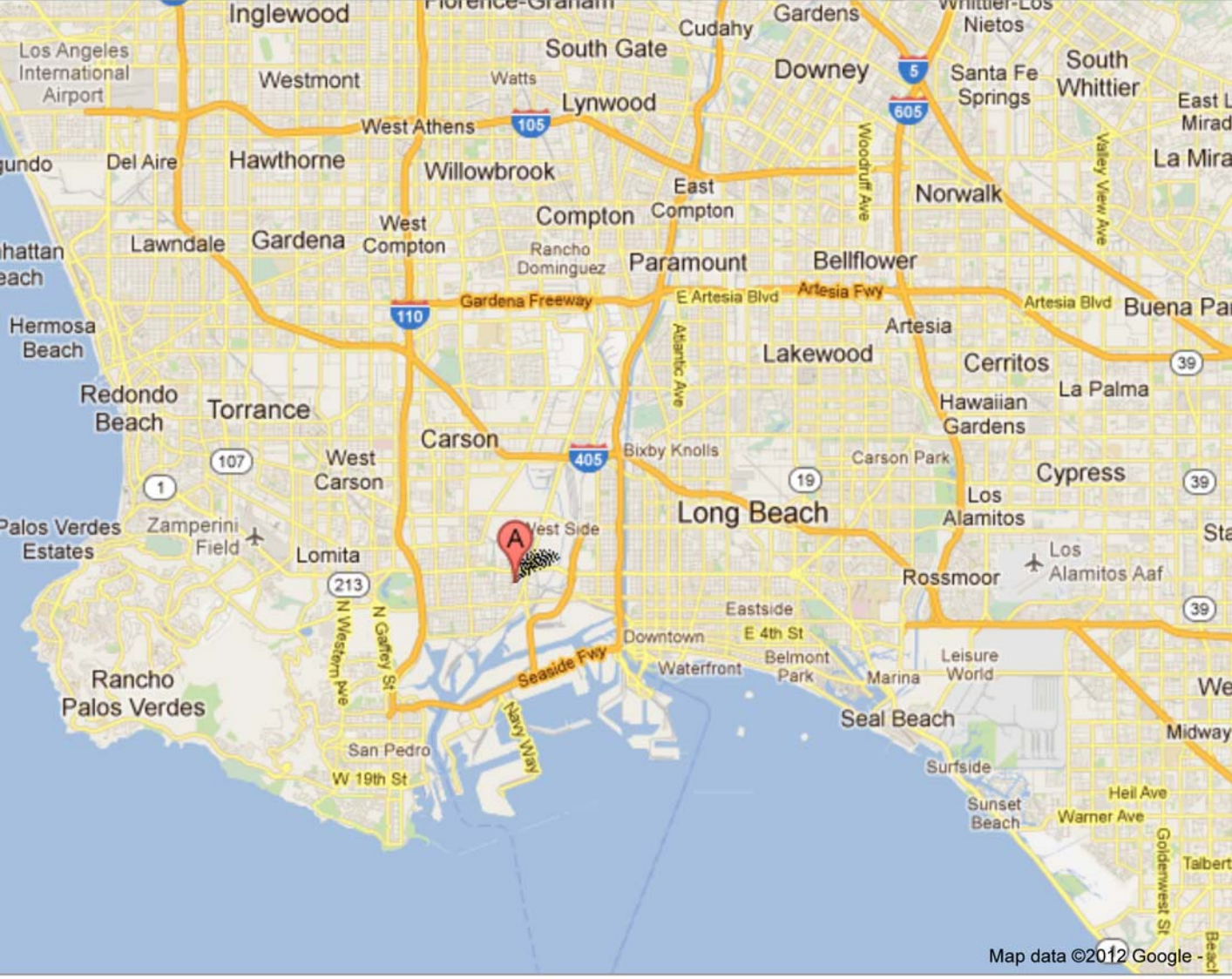
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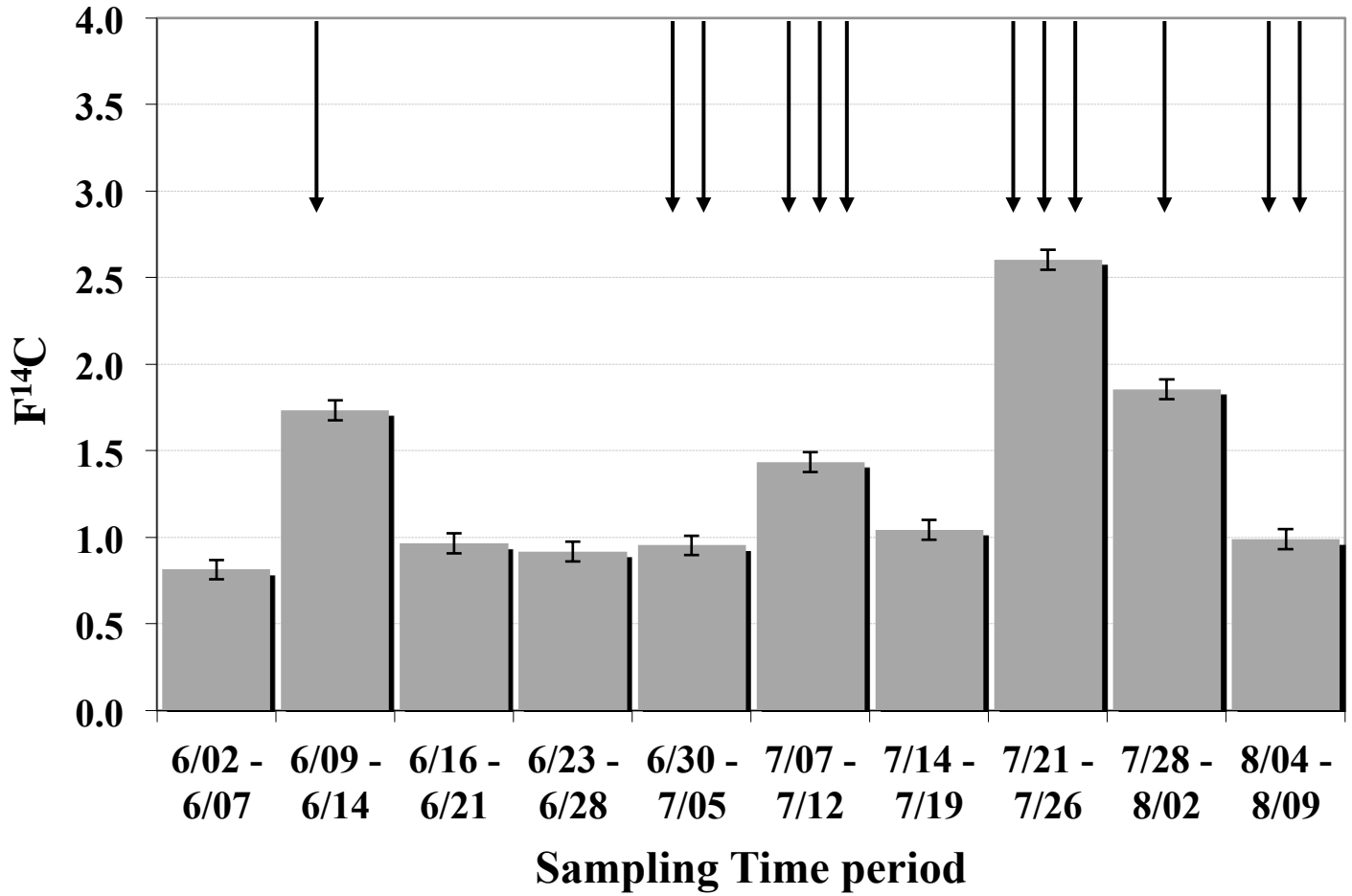
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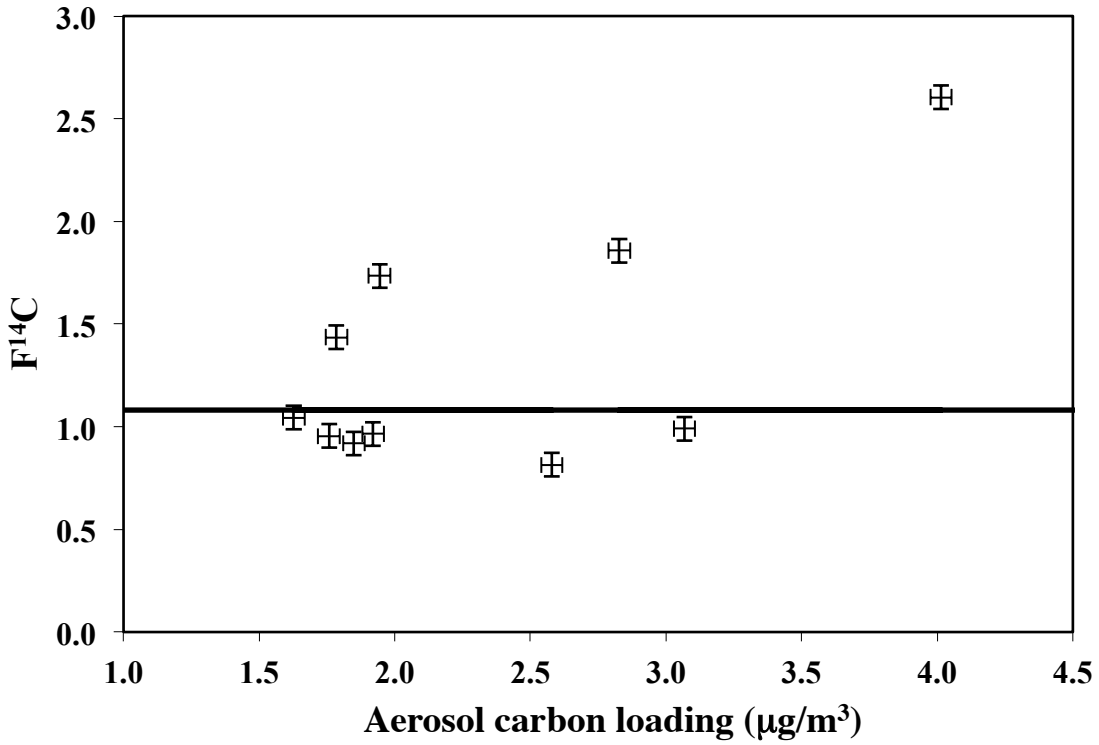
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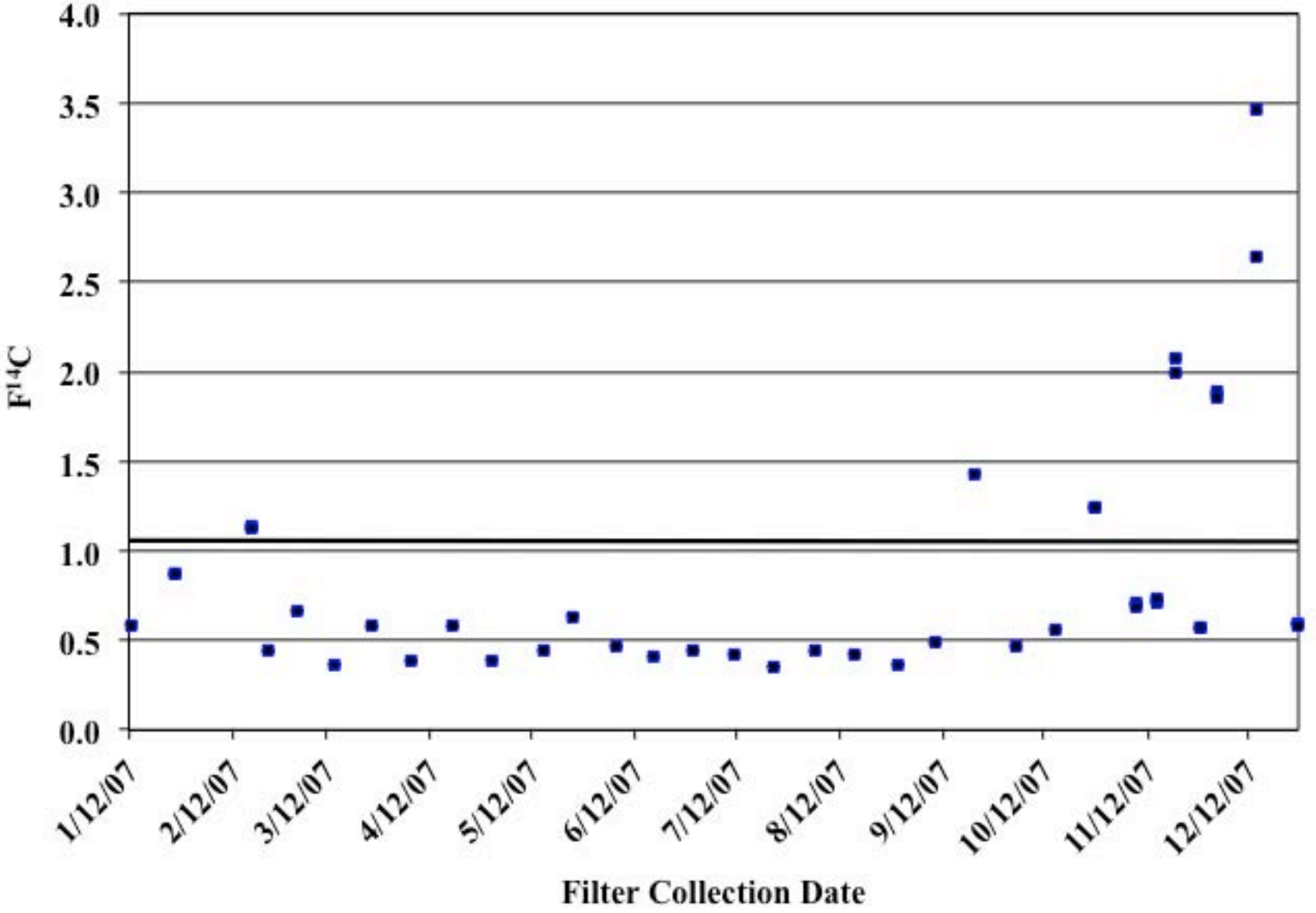
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